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Effects of a Nonlinear Induced Electric Dipole Moment at 1w

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To: Nonlinear Propagators
From: John Trenholme
Subject: Effects of a Nonlinear Induced Electric Dipole Moment at 1ω

This version has WFW {eq} codes in the equations

Summary

This memo begins with a survey of the present knowledge of nonlinear propagation effects at 1ω in isotropic materials (i.e., glasses) due to an induced electric dipole moment which has a small nonlinear field dependence. The important effects are phase retardation, ellipse rotation, and small-scale ripple growth. The analysis is then extended to the case of strong beams at a large angle, as in a zig-zag slab amplifier.

Maxwell's Equations in a Nonlinear Medium

We follow the usual electromagnetic wave treatment here, but take care to allow for the possibility of a nonlinear component to the induced electric dipole moment per unit volume. To maintain sanity, we limit the complexity of the resulting expressions by considering only stationary isotropic media, including isotropy of the nonlinear response. This will work for propagation in unstressed glass, but we will have to do the real job in crystals (even cubic crystals like NaCl, since they can and often do have anisotropic nonlinear response). Units are rationalized MKS (meaning no 4π 's will appear).

Let's get an equation for the electric field. We start with the Maxwell equation

$$\nabla \times \underline{\mathbf{E}} = - \frac{\partial \underline{\mathbf{B}}}{\partial t}$$

and take its curl to get

$$\nabla \times \nabla \times \underline{\mathbf{E}} = - \frac{\partial}{\partial t} (\nabla \times \underline{\mathbf{B}}) .$$

We now have to introduce the magnetic response of the material to the fields. We assume it to be linear, instantaneous and isotropic, so that

$$\underline{\mathbf{B}} = \mu_0 (\underline{\mathbf{H}} + \underline{\mathbf{M}}) = \mu \underline{\mathbf{H}} .$$

The most likely case is that the material is nonmagnetic so $\underline{\mathbf{M}} = 0$ and therefore $\mu = \mu_0$. We now have

$$\nabla \times \nabla \times \underline{\mathbf{E}} = -\mu \frac{\partial}{\partial t} (\nabla \times \underline{\mathbf{H}}) .$$

Another of Maxwell's gems can be used to simplify $\nabla \times \underline{\mathbf{H}}$, which is equal to the sum of the real and displacement currents:

$$\nabla \times \underline{\mathbf{H}} = \underline{\mathbf{J}} + \frac{\partial \underline{\mathbf{D}}}{\partial t} .$$

We dismiss the possibility of any real current $\underline{\mathbf{J}}$ by forbidding source currents and requiring the material's conductivity σ to be zero, so that

$$\sigma \underline{\mathbf{E}} = 0$$

which leaves us with the displacement current $\partial \underline{\mathbf{D}} / \partial t$ as the only current. We now have

$$\nabla \times \nabla \times \underline{\mathbf{E}} = -\mu \frac{\partial}{\partial t} \left(\frac{\partial \underline{\mathbf{D}}}{\partial t} \right) .$$

At this point, the induced electric dipole of the material comes on stage. We recall that

$$\underline{\mathbf{D}} = \epsilon_0 \underline{\mathbf{E}} + \underline{\mathbf{P}}$$

where $\underline{\mathbf{P}}$ is the electric dipole moment per unit volume in the material. In a linear isotropic dielectric with instantaneous response the only moment would be that induced by the field (no electrets) and we would have the relation

$$\underline{\mathbf{P}} = \epsilon_0 \chi \underline{\mathbf{E}}$$

so that

$$\underline{\mathbf{D}} = \epsilon_0 (1 + \chi) \underline{\mathbf{E}} = \epsilon \underline{\mathbf{E}}$$

with χ (and therefore ϵ) a scalar constant. In a nonlinear dielectric, on the other hand, χ depends on the local field $\underline{\mathbf{E}}$ (more on this later) and so χ and ϵ are not constant in either space or time. We will, however, assume that the nonlinear moment depends on the instantaneous local field $\underline{\mathbf{E}}$, just as we assumed the linear moment did.

Returning to Maxwell, we have

$$\nabla \times \nabla \times \underline{\mathbf{E}} = -\mu \frac{\partial}{\partial t} \left(\frac{\partial \underline{\mathbf{D}}}{\partial t} \right) = -\mu \frac{\partial^2}{\partial t^2} (\epsilon \underline{\mathbf{E}})$$

and we simplify this by use of the identity

$$\nabla \times \nabla \times \underline{\mathbf{E}} = \nabla (\nabla \cdot \underline{\mathbf{E}}) - \nabla^2 \underline{\mathbf{E}}$$

to get

$$\nabla^2 \underline{\mathbf{E}} = \mu \frac{\partial^2}{\partial t^2} (\epsilon \underline{\mathbf{E}}) + \nabla (\nabla \cdot \underline{\mathbf{E}}) .$$

Note that ∇^2 on a vector is not the same as ∇^2 on a scalar, although they are similar in rectangular coordinates.

We deal with the $\nabla \cdot \underline{\mathbf{E}}$ term by the use of Maxwell's equation

$$\nabla \cdot \underline{\mathbf{D}} = \rho$$

and proceed to summarily forbid free charge in the same autocratic way in which we refused to consider current sources. Then we have (recall that $\underline{\mathbf{D}} = \epsilon \underline{\mathbf{E}}$)

$$\nabla \cdot \underline{\mathbf{D}} = 0 = \nabla \cdot (\epsilon \underline{\mathbf{E}}) = \epsilon \nabla \cdot \underline{\mathbf{E}} + \underline{\mathbf{E}} \cdot \nabla \epsilon$$

so that

$$\nabla \cdot \underline{\mathbf{E}} = -\frac{\underline{\mathbf{E}} \cdot \nabla \epsilon}{\epsilon}$$

which shows that $\nabla \cdot \underline{\mathbf{E}}$ is non-zero in a charge-free material only if ϵ varies in space. For an isotropic material, this happens only in the presence of a nonlinearity. In fact, if we separate ϵ into its vacuum, linear dielectric and nonlinear dielectric parts we have

$$\epsilon = \epsilon_0(1+\chi) = \epsilon_0(1+\chi_L+\chi_N)$$

and see that the only part that varies in space and gives a contribution to the gradient is the χ_N part. We then have

$$\nabla \cdot \mathbf{E} = - \frac{\mathbf{E} \cdot \nabla \chi_N}{1 + \chi} .$$

By similar reasoning, we can rewrite the second time derivative of $\epsilon \mathbf{E}$ in terms of the linear and nonlinear parts as

$$\frac{\partial^2}{\partial t^2} (\epsilon \mathbf{E}) = \epsilon_0 \left[(1 + \chi) \frac{\partial^2 \mathbf{E}}{\partial t^2} + \frac{\partial^2}{\partial t^2} (\chi_N \mathbf{E}) \right] .$$

Our final result is an equation for \mathbf{E} which incorporates Maxwell's equations and allows for the existence of an induced electric dipole moment which depends nonlinearly on the applied field. We write it in a form which has the linear terms on the left and the nonlinear terms on the right:

$$\boxed{\nabla^2 \mathbf{E} - \mu \epsilon_L \frac{\partial^2 \mathbf{E}}{\partial t^2} = \mu \epsilon_0 \frac{\partial^2}{\partial t^2} (\chi_N \mathbf{E}) - \nabla \left(\frac{\mathbf{E} \cdot \nabla \chi_N}{1 + \chi} \right)} .$$

For convenience, we have used the definition

$$\epsilon_L = \epsilon_0 (1 + \chi_L) .$$

Propagating Waves

Now (at last!) we can propagate some waves. First, consider the solutions of the linear equation (which we get by setting the right-hand side of the wave equation to zero)

$$\nabla^2 \mathbf{E} = \mu \epsilon_L \frac{\partial^2 \mathbf{E}}{\partial t^2} .$$

This is known as the vector Helmholtz equation. It's easy to show¹ that a general solution to this equation is

$$\mathbf{E} = \hat{\mathbf{p}} f \left(t - \hat{\mathbf{u}} \cdot \mathbf{r} \sqrt{\mu \epsilon_L} \right)$$

where $f()$ is an arbitrary function, $\hat{\mathbf{u}}$ is a unit vector in any direction, and $\hat{\mathbf{p}}$ is a unit polarization vector perpendicular to $\hat{\mathbf{u}}$. These solutions are plane waves moving in the $\hat{\mathbf{u}}$ direction. They

¹ see, for example, section 11-2 of CLASSICAL ELECTRICITY AND MAGNETISM by Panofsky and Phillips

have no variation in the plane perpendicular to \hat{u} , and move at a constant velocity $v = 1/\sqrt{\mu\epsilon_L}$. It is one of the famous triumphs of physics that if we use the vacuum values μ_0 and ϵ_0 we find this velocity is the speed of light

$$v = c = \frac{1}{\sqrt{\mu_0\epsilon_0}} .$$

Inside a material, this velocity is changed to the velocity

$$v = \frac{1}{\sqrt{\mu\epsilon_L}} = c \sqrt{\frac{\mu_0\epsilon_0}{\mu\epsilon_L}} = \frac{c}{n}$$

where n is the refractive index, defined as

$$n = \sqrt{\frac{\mu\epsilon_L}{\mu_0\epsilon_0}} = \sqrt{\frac{\mu(1+\chi_L)}{\mu_0}} .$$

In a non-magnetic medium $\mu = \mu_0$ and

$$n = \sqrt{1+\chi_L} .$$

For glasses used in lasers, we recall that $n \cong 1.4$ so the numerical magnitude of χ_L is about unity in such glasses. Note that the reason for a reduced velocity in a material is the addition of an induced electric dipole moment in phase with the vacuum moment $\epsilon_0 \underline{E}$.

In a stationary, isotropic, linear, charge-free, source-current-free, nonconducting, nondispersive medium (commonly referred to as "free space", although we here allow the presence of a linear dielectric), the total electromagnetic field can always be expanded as a sum of these plane waves with two orthogonal polarization vectors \hat{p} for each direction \hat{u} . Since the governing equation is linear, these waves do not interact. In the presence of a nonlinearity, the propagation velocity of waves is changed, and waves are coupled. We will only consider cases where these velocity changes and wave mixings are small.

Sinusoidal Waves

For lasers, we are interested in waves narrowly spaced around one frequency. Restricting our attention to near-monochromatic sinusoidal waves leads to considerable simplification. At any point in space, a sinusoidal wave oscillates as

$$\sin(\omega t - \phi)$$

where ω is the angular frequency (radians per second) and ϕ is a constant phase. We have chosen to use the trigonometric form here rather than the more common complex exponential form because of the well-known problems with the evaluation of powers of fields when using complex exponentials. We now write the general propagating wave above as

$$f(t - \hat{\mathbf{u}} \cdot \mathbf{r} \sqrt{\mu\epsilon_L}) = \sin(\omega t - \mathbf{k} \cdot \mathbf{r} - \phi)$$

by multiplying the argument of the function through by ω . When we do that, we find that the magnitude of the propagation vector \mathbf{k} is given by

$$|\mathbf{k}| = k = \frac{\omega}{v} = \frac{2\pi}{\lambda} = \frac{2\pi n}{\lambda_v}$$

where λ is the wavelength in the material and λ_v is the wavelength in vacuum. The general monochromatic wave is therefore of the form

$$\mathbf{E} = \hat{\mathbf{p}} A \sin(\omega t - \mathbf{k} \cdot \mathbf{r} - \phi)$$

and any monochromatic field can be expanded with two orthogonal polarizations and two orthogonal phases per \mathbf{k} , and with \mathbf{k} values pointing in all possible directions.

With this form of the wave, we have the simple result

$$\nabla^2 \mathbf{E} = -k^2 \mathbf{E}$$

for the space derivative in the linear plane wave equation. Recall that k is a scalar equal to the magnitude of \mathbf{k} . The time derivative becomes

$$\mu\epsilon_L \frac{\partial^2 \mathbf{E}}{\partial t^2} = -\omega^2 \mu\epsilon_L \mathbf{E}$$

and the wave equation is

$$\boxed{[\omega^2 \mu\epsilon_L - k^2] \mathbf{E} = \mu\epsilon_0 \frac{\partial^2}{\partial t^2} (\chi_N \mathbf{E}) - \nabla \left(\frac{\mathbf{E} \cdot \nabla \chi_N}{1 + \chi} \right)} .$$

Once we are monochromatic, we can relax the assumption of constant response at all frequencies ("instantaneous response") and allow ϵ to depend on the frequency ω that we are using. Allowing such a dependence will permit us to do problems with dispersion.

The Local Electric Field

In any volume much smaller than a wavelength but large enough to contain many atoms, we can always find a plane in which an arbitrary sum of monochromatic applied fields lies, and in which that sum of fields is composed of right circular and left circular polarization components

$$\underline{\mathbf{E}} = C \hat{\mathbf{e}}_R + D \hat{\mathbf{e}}_L$$

where the polarization basis vectors are given by

$$\hat{\mathbf{e}}_R = \frac{\hat{\mathbf{a}} \cos(\omega t) + \hat{\mathbf{b}} \sin(\omega t)}{\sqrt{2}}$$

$$\hat{\mathbf{e}}_L = \frac{\hat{\mathbf{a}} \cos(\omega t) - \hat{\mathbf{b}} \sin(\omega t)}{\sqrt{2}} \quad .$$

The unit vectors $\hat{\mathbf{a}}$ and $\hat{\mathbf{b}}$ lie in the field plane and are at right angles. To see how we can always get this form for the local field, add up the fields along $\hat{\mathbf{x}}$, $\hat{\mathbf{y}}$ and $\hat{\mathbf{z}}$ due to all \mathbf{k} and $\hat{\mathbf{p}}$ and ϕ components of all the waves in the field. The general result will be

$$\underline{\mathbf{E}} = \hat{\mathbf{x}} [E_{xc} \cos(\omega t) + E_{xs} \sin(\omega t)] \\ + \hat{\mathbf{y}} [E_{yc} \cos(\omega t) + E_{ys} \sin(\omega t)] \\ + \hat{\mathbf{z}} [E_{zc} \cos(\omega t) + E_{zs} \sin(\omega t)] \quad .$$

We now add up the cosine and sine parts separately to get

$$\underline{\mathbf{E}} = \underline{\mathbf{E}}_C \cos(\omega t) + \underline{\mathbf{E}}_S \sin(\omega t)$$

where the vectors associated with cosine and sine are

$$\underline{\mathbf{E}}_C = \hat{\mathbf{x}} E_{xc} + \hat{\mathbf{y}} E_{yc} + \hat{\mathbf{z}} E_{zc}$$

and

$$\underline{\mathbf{E}}_S = \hat{\mathbf{x}} E_{xs} + \hat{\mathbf{y}} E_{ys} + \hat{\mathbf{z}} E_{zs} \quad .$$

Since $\underline{\mathbf{E}}$ is formed from a linear combination of two vectors, it must always lie in the plane defined by those two vectors. Therefore, the most general monochromatic field is locally planar.

We put the field into circular polarization form by separately decomposing the linearly polarized cosine and sine parts, and adding them. Since a linear polarization is a sum of equal parts right and left circular polarization, we have

$$\underline{\mathbf{E}}_C = \frac{1}{2} (\hat{\mathbf{e}}_R + \hat{\mathbf{e}}_L)$$

The local field above is not in the form of an elliptic polarization unless $\underline{\mathbf{E}}_S$ is perpendicular to $\underline{\mathbf{E}}_C$, which is usually not the case. We can, however, always find a set of orthogonal axes in the field plane which do give elliptic polarization. In the plane defined by $\underline{\mathbf{E}}_S$ and $\underline{\mathbf{E}}_C$, introduce orthogonal coordinate vectors $\hat{\mathbf{a}}$ and $\hat{\mathbf{b}}$. Find the components of $\underline{\mathbf{E}}_S$ and $\underline{\mathbf{E}}_C$ along these vectors and write

$$\begin{aligned} \underline{\mathbf{E}} = & \hat{\mathbf{a}} [\hat{\mathbf{a}} \cdot \underline{\mathbf{E}}_S \sin(\omega t) + \hat{\mathbf{a}} \cdot \underline{\mathbf{E}}_C \cos(\omega t)] \\ & + \hat{\mathbf{b}} [\hat{\mathbf{b}} \cdot \underline{\mathbf{E}}_S \sin(\omega t) + \hat{\mathbf{b}} \cdot \underline{\mathbf{E}}_C \cos(\omega t)] . \end{aligned}$$

If these components are to be in quadrature, the electric field must have the form

$$\underline{\mathbf{E}} = C_1 \hat{\mathbf{a}} \sin(\omega t + \zeta) + C_2 \hat{\mathbf{b}} \cos(\omega t + \zeta)$$

where ζ is a common coordinate rotation angle chosen to put $\underline{\mathbf{E}}$ in this form. We can expand this to

$$\begin{aligned} \underline{\mathbf{E}} = & C_1 \hat{\mathbf{a}} [\sin(\omega t) \cos(\zeta) + \cos(\omega t) \sin(\zeta)] \\ & + C_2 \hat{\mathbf{b}} [\cos(\omega t) \cos(\zeta) - \sin(\omega t) \sin(\zeta)] . \end{aligned}$$

Comparing the two forms for $\underline{\mathbf{E}}$, we see that we must have

$$\tan(\zeta) = \frac{\hat{\mathbf{a}} \cdot \underline{\mathbf{E}}_C}{\hat{\mathbf{a}} \cdot \underline{\mathbf{E}}_S} = - \frac{\hat{\mathbf{b}} \cdot \underline{\mathbf{E}}_S}{\hat{\mathbf{b}} \cdot \underline{\mathbf{E}}_C} .$$

If we define α as the angle from $\hat{\mathbf{a}}$ to $\underline{\mathbf{E}}_S$ and β as the angle from $\underline{\mathbf{E}}_S$ to $\underline{\mathbf{E}}_C$, then the above relationship becomes

$$\underline{\mathbf{E}}_{\mathbf{C}} \cdot \underline{\mathbf{E}}_{\mathbf{C}} \cos(\alpha+\beta) \sin(\alpha+\beta) + \underline{\mathbf{E}}_{\mathbf{S}} \cdot \underline{\mathbf{E}}_{\mathbf{S}} \cos(\alpha) \sin(\alpha) = 0$$

which solves to give

$$\alpha = \frac{1}{2} \tan^{-1} \left(\frac{\sin(2\beta)}{\frac{\underline{\mathbf{E}}_{\mathbf{S}} \cdot \underline{\mathbf{E}}_{\mathbf{S}}}{\underline{\mathbf{E}}_{\mathbf{C}} \cdot \underline{\mathbf{E}}_{\mathbf{C}}} + \cos(2\beta)} \right) .$$

We find $\cos(\beta)$ from the normalized dot product of $\underline{\mathbf{E}}_{\mathbf{C}}$ and $\underline{\mathbf{E}}_{\mathbf{S}}$, and get $\cos(2\beta)$ and $\sin(2\beta)$ from $\cos(\beta)$. This demonstrates by construction that the local field is always planar and has elliptical polarization, a fact which will greatly simplify the next section.

Nonlinear Induced Electric Dipole Moment

In a nonlinear medium, the induced electric dipole moment has an added component which depends nonlinearly on the local electric field. If the medium is isotropic, the induced moment must lie in the direction of the applied field, and have a magnitude which depends only on the magnitude, but not the direction, of the applied field. It must therefore depend only on $\underline{\mathbf{E}}$ times the dot product $\underline{\mathbf{E}} \cdot \underline{\mathbf{E}}$, or on $\underline{\mathbf{E}}$ times integer powers of $\underline{\mathbf{E}} \cdot \underline{\mathbf{E}}$. We assume the induced electric dipole moment $\underline{\mathbf{P}}$ is mostly linear in the applied field, and expand it in powers of $\underline{\mathbf{E}} \cdot \underline{\mathbf{E}}$ in the form

$$\underline{\mathbf{P}} = \epsilon_0 [\chi_1 + \chi_3 \underline{\mathbf{E}} \cdot \underline{\mathbf{E}} + \chi_5 (\underline{\mathbf{E}} \cdot \underline{\mathbf{E}})^2 + \dots] \underline{\mathbf{E}}$$

where $\epsilon_0 \chi_1 \underline{\mathbf{E}}$ is the usual linear induced moment. Ignoring terms beyond χ_3 as too small to matter, we concentrate on the $\chi_3 \underline{\mathbf{E}} \cdot \underline{\mathbf{E}}$ term. Note that χ_1 is unitless, but χ_3 has the units [field⁻²]. From the previous section, we know that the local field is of the form

$$\underline{\mathbf{E}} = A \hat{\mathbf{a}} \sin(\omega t) + B \hat{\mathbf{b}} \cos(\omega t)$$

where $\hat{\mathbf{a}}$ and $\hat{\mathbf{b}}$ are at right angles. Then the dot product is

$$\underline{\mathbf{E}} \cdot \underline{\mathbf{E}} = A^2 \sin^2(\omega t) + B^2 \cos^2(\omega t)$$

and so the moment has the form

$$\begin{aligned} \underline{\mathbf{P}} = & \epsilon_0 \chi_1 \underline{\mathbf{E}} \\ & + \epsilon_0 \chi_3 \hat{\mathbf{a}} [A^3 \sin^3(\omega t) + AB^2 \sin(\omega t) \cos^2(\omega t)] \\ & + \epsilon_0 \chi_3 \hat{\mathbf{b}} [A^2 B \sin^2(\omega t) \cos(\omega t) + B^3 \cos^3(\omega t)] . \end{aligned}$$

This somewhat unpleasant result can be simplified by rejecting the harmonic terms. From trigonometry, we have

$$\begin{aligned}\sin^3 \theta &= \frac{3 \sin \theta - \sin 3\theta}{4} \\ \sin \theta \cos^2 \theta &= \frac{\sin \theta + \sin 3\theta}{4} \\ \sin^2 \theta \cos \theta &= \frac{\cos \theta - \cos 3\theta}{4} \\ \cos^3 \theta &= \frac{3 \cos \theta + \cos 3\theta}{4}\end{aligned}$$

from which we see that the nonlinear induced moment has components at ω and at 3ω . We assume that the 3ω components are not phase-matched, and therefore do not affect our results (more on phase-matching later). This means we need to keep only the 1ω parts. The moment at 1ω is

$$\mathbf{P} = \epsilon_0 \chi_1 \mathbf{E} + \epsilon_0 \chi_3 \left[\left(\frac{3A^2 + B^2}{4} \right) A \hat{a} \sin(\omega t) + \left(\frac{A^2 + 3B^2}{4} \right) B \hat{b} \cos(\omega t) \right]$$

which can be written as

$$\begin{aligned}\mathbf{P} = \epsilon_0 \left\{ \left[\chi_1 + \frac{3}{4} \chi_3 (A^2 + B^2) \right] \mathbf{E} \right. \\ \left. + \frac{1}{4} \chi_3 [B^2 A \hat{a} \sin(\omega t) + A^2 B \hat{b} \cos(\omega t)] \right\} .\end{aligned}$$

We can understand this by considering some special cases. First, consider the case of linear polarization, which has $A \neq 0$ and $B = 0$. The moment becomes simply

$$\mathbf{P} = \epsilon_0 \left(\chi_1 + \frac{3}{4} \chi_3 A^2 \right) \mathbf{E} .$$

Next, consider circular polarization, where $A = B$. We require the circularly polarized field to have the same energy density (locally) as the linearly polarized field. This energy density is proportional to the time average of $\mathbf{E} \cdot \mathbf{E}$, which is $(A^2 + B^2)/2$, so we must have both components reduced by the square root of 2. Then in terms of the coefficient A of the linear field, the induced moment is

$$\underline{\mathbf{P}} = \epsilon_0 \left[\chi_1 + \frac{1}{4} \chi_3 \left(\frac{A^2}{2} + \frac{A^2}{2} \right) \right] \underline{\mathbf{E}} = \epsilon_0 \left(\chi_1 + \frac{1}{2} \chi_3 A^2 \right) \underline{\mathbf{E}} .$$

This induced moment is 2/3 of the amount due to linearly polarized light of the same local intensity.

The general case is found by use of the full formula. The main point to note is that when we use linear polarizations as our basis, each polarization of the induced nonlinear dipole moment has a component proportional to the intensity of the applied field of that polarization, and also a component proportional to one-third the intensity in the quadrature polarization. It is therefore incorrect to say that the applied electric field raises the refractive index, since there is a cross-coupling of polarization components as well.

It is interesting to change our polarization basis from linear to circular. The base polarizations become right and left circular, with the general field being given by

$$\underline{\mathbf{E}} = C \left\{ \frac{\hat{a} \sin(\omega t) + \hat{b} \cos(\omega t)}{\sqrt{2}} \right\} + D \left\{ \frac{\hat{a} \sin(\omega t) - \hat{b} \cos(\omega t)}{\sqrt{2}} \right\} .$$

We write this as

$$\underline{\mathbf{E}} = C \hat{\mathbf{e}}_R + D \hat{\mathbf{e}}_L$$

for compactness.

The factors of $\sqrt{2}$ are used so that $A = 1$ and $C = 1$ (for example) will have the same local energy density. With these basis vectors, the induced dipole moment at 1ω can be written as

$$\begin{aligned} \underline{\mathbf{P}} = & \epsilon_0 \chi_1 \underline{\mathbf{E}} + \epsilon_0 \chi_3 \left[\left(\frac{C^2 + 2D^2}{2} \right) C \hat{\mathbf{e}}_R + \left(\frac{2C^2 + D^2}{2} \right) D \hat{\mathbf{e}}_L \right] \\ & + \epsilon_0 \left\{ \left[\chi_1 + \frac{1}{2} \chi_3 (C^2 + D^2) \right] \underline{\mathbf{E}} + \frac{1}{2} \chi_3 (D^2 C \hat{\mathbf{e}}_R + C^2 D \hat{\mathbf{e}}_L) \right\} \end{aligned}$$

which shows that the opposite circular polarization has *twice* the effect on a circular polarization component as the effect of that circular polarization on itself. This is because $\underline{\mathbf{E}} \cdot \underline{\mathbf{E}}$ is constant in time for pure circular polarization, but any small addition of the opposite circular polarization causes temporal modulation of ϵ .

Propagation of a Single Plane Wave

Let us now consider the propagation of a single plane wave in the presence of a nonlinearity. We assume that the electric field is of the plane-wave elliptic form

$$\mathbf{E} = A \hat{\mathbf{a}} \sin(\omega t - \mathbf{k} \cdot \mathbf{r}) + B \hat{\mathbf{b}} \cos(\omega t - \mathbf{k} \cdot \mathbf{r})$$

where we have dropped a constant phase ϕ by redefining the origin of time. Note that we may have to rotate our coordinate system around \mathbf{k} in order to get this pure elliptic form, just as in the local electric field section. Since $\nabla \chi_N$ is along \mathbf{k} for this field, $\mathbf{E} \cdot \nabla \chi_N$ is zero and the only nonlinear term is the second derivative with respect to time. Also, the local electric field is the same everywhere (except for a trivial phase factor) so we can write the wave equation for the 1ω field as

$$\begin{aligned} [\omega^2 \mu \epsilon_L - k^2] \mathbf{E} &= [\omega^2 \mu \epsilon_0 (1 + \chi_L) - k^2] \mathbf{E} = \\ &= -\omega^2 \mu \epsilon_0 \chi_3 \left[\left(\frac{3A^2 + B^2}{4} \right) A \hat{\mathbf{a}} \sin(\omega t - \mathbf{k} \cdot \mathbf{r}) + \left(\frac{A^2 + 3B^2}{4} \right) B \hat{\mathbf{b}} \cos(\omega t - \mathbf{k} \cdot \mathbf{r}) \right]. \end{aligned}$$

throughout all of space.

As before, we first consider the case of linear polarization with $A \neq 0$ and $B = 0$. We get

$$[\omega^2 \mu \epsilon_0 (1 + \chi_L + \frac{3}{4} \chi_3 A^2) - k^2] \mathbf{E} = 0$$

which gives a propagation velocity of

$$v = \frac{\omega}{k} = \frac{1}{\sqrt{\mu \epsilon_0 (1 + \chi_L + \frac{3}{4} \chi_3 A^2)}} = \frac{v_L}{\sqrt{1 + \frac{3 \chi_3 A^2}{4(1 + \chi_L)}}}$$

where v_L is the velocity with only linear dielectric response

$$v_L = \frac{1}{\sqrt{\mu \epsilon_0 (1 + \chi_L)}}.$$

We have found a self-consistent solution in which the wave remains a plane wave, but in which its velocity of propagation is changed. Under our assumption of small nonlinear effects, we can expand the square root to write this velocity as

$$v \cong v_L \left(1 - \frac{3\chi_3 A^2}{8(1+\chi_L)} \right).$$

If we propagate the wave a distance L at this velocity, its phase (in radians) will be different from that of a wave propagating at a velocity v_L by the amount

$$B = (k - k_L)L \cong \frac{\omega(v_L - v)L}{v_L^2} = \frac{\omega}{v_L} \frac{3\chi_3 A^2 L}{8(1+\chi_L)} = \frac{3\pi\mu\chi_3 A^2 L}{4\lambda_V \mu_0 n}.$$

This phase delay is the famous B integral. In terms of the usual nonlinear coefficient γ of the material, the B integral for a single plane wave of one polarization is defined by

$$B = \frac{2\pi}{\lambda_V} \int_0^L \gamma I \, ds$$

where I is the intensity of the wave.

Propagation of Two Near-Collinear Waves

----- Not ready for prime time -----

Propagation of Two Waves at a Large Angle

----- Not ready for prime time -----